INVESTIGATION OF COHERENT SOURCES OF INFRARED RADIATION

under the direction of

A. L. Schawlow

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Microwave Laboratory
W. W. Hansen Laboratories of Physics
Stanford   University
Stanford, California

STAFF

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PRINCIPAL INVESTIGATOR

A. L. Schawlow, Professor

RESEARCH ASSISTANT

B. McCaul

INTRODUCTION

This program is concerned with new methods of generating and detecting far infrared radiation and with their applications to problems of physical interest. The over-all purpose is to advance the technology of the infrared region so that it may become as accessible for scientific investigations as the radio and optical portions of the spectrum.

PRESENT STATUS

Investigation has begun with consideration of the CN radical as the active species of a coherent infrared source. It is hoped that the stimulated emission observed arises between states which will show a net shift in a magnetic field and thus provide a tunable infrared source. A number of molecular gases have shown laser action in the far infrared region. The effects observed with CN will give some indication of the possibilities and problems to be expected with other gases.

The laser line state assignments and population inversion mechanism are currently being discussed in the literature. Chantry et al.,[1] established the CN radical as the radiating species. In explaining the emission at 337 μ, they pointed to a transition of the appropriate energy in the electronic ground state $X^2\Sigma(v = 2$, $K = 8) \rightarrow X^2\Sigma(2,7)$. They cited the rotational perturbation of $B^2\Sigma(0,7)$ and $A^2\pi(10,7)$ as the mechanism permitting the red system transition $A^2\pi \rightarrow X^2\Sigma$ to go as favorably as the violet $B^2\Sigma \rightarrow X^2\Sigma$, thus overpopulating the $X^2\Sigma(2,8)$ state.

Broida et al.,[2] pointed out that if this inversion mechanism were correct many other inversions and inversion mechanisms would seem likely. They suggested as examples the $K = 4$, 11, and 15 perturbed levels of the $B^2\Sigma$, $v = 0$ and $A^2\pi$, $v = 10$ bands; the R branch transition of the $K = 7$ perturbation; and the perturbations between rotational levels of $X^2\Sigma$, $v = 11$ and $A^2\pi$, $v = 7$. They suggested that inversions would be more likely in the $v = 0$ and 1 bands rather than the $v = 2$ band of the electronic ground state. Finally, they made estimates

from rate equations which said that the Chantry mechanism was improbable. However, by this time Mathias et al.,[3] had found other weaker laser transitions in CN systems.

Stefan et al.,[4] at this time using a 6.4 m laser tube, reported a splitting of the 337 μ line plus new emissions in cyanic compounds at 310, 537, and 538 μ. They attributed these to rotational transitions in the $v = 2$ and $v = 16$ bands of the ground state on the basis of wavelength coincidence, and they cited Broida et al.,[2] for the inversion mechanisms. Then Stefan et al.,[5] found stimulated emission at 724 μ in ICN for which they found no wavelength coincidence in the ground rotational levels. By adjusting the vibrational terms of the data of Jenkins et al.,[6] by 1.06 cm^{-1}, an amount which did not exceed the uncertainty in the vibrational term measurement, they were able to find new coincidences in wavelength for all the observed levels. These new assignments were all transitions between electronic levels associated with the rotational perturbation between $X^2\Sigma$, $v = 11$ and $A^2\pi$, $v = 7$; the assignments failed to account for the observed splittings.

Recently Stefan et al.,[7] have published observation of another line from ICN at 676 μ and a third set of assignments for all the observed transitions, claiming that they had previously demonstrated that the laser emissions were not explained by current theories. These assignments are all transitions from the $A^2\pi$ state in which "CN radicals are formed almost exclusively" to $B^2\Sigma$ in the case of the strong 337 and 310 μ emission and to $X^2\Sigma$ in the case of the five other weaker emissions. No rotationally perturbed levels are involved. They cite the observation of Brown and Broida[8] of strong violet transitions from the lower laser

level of the stronger emissions. Frequency coincidences range from

-0.07 to +0.14 cm^{-1}; instrumental accuracy was better than this. No

mention is made of previously cited small splitting at 337 μ and 538 μ.

The question then of tunability in a magnetic field is closely

connected with the nature of the energy levels involved in laser action.

If all the laser transitions occur between rotational levels of the

ground state where spin doubling is small, one expects in moderate

fields ($\sim$ 1000 gauss, depending on K) a decoupling of the electronic

spin from the rotational angular momentum K . For all K in a $^2\Sigma$

state each rotational level is split effectively into two components

each having 2K + 1 degeneracy corresponding to $m_s = \pm 1/2$. While

these levels are known to shift by about 1 cm^{-1} in a 10 kilogauss field,[9]

the selection rules $\Delta m_s = 0$ for the strongest transitions imply that

there would be no net shift in the laser transition. In low magnetic

fields K is coupled to the spin so that there would be a net shift

corresponding to:[10]

$$\Delta W_{J=N+1/2} = \frac{1.001 \, M\mu_0 H}{J}$$

$$\Delta W_{J=N-1/2} = - \frac{1.001 \, M\mu_0 H}{J + 1} \quad .$$

A similar situation obtains for any $B^2\Sigma \rightarrow X^2\Sigma$ transition; these bands

show no magnetic effect in a magnetic field except near perturbations

and except for doublet narrowing.

For $A^2\pi \to {}^2\Sigma$ transitions the situation would be quite different, the most obvious difference being that while $A^2\pi_{1/2}$ states of the third Stefan et al.,[7] upper level laser transition assignment have only a small Zeeman shift, the lower ${}^2\Sigma$ states would shift by a large amount.

Thus there are interesting possibilities for a magnetically-tuned CN laser. It is clear to us that further elucidation of the population inversion and stimulated emission kinetics is desirable. We will have three experimental approaches to this question. First, we expect to have data on the Zeeman behavior of the laser emission. Second we will be able to study at optical and infrared frequencies the time-dependent behavior of levels important to proposed laser excitation and relaxation mechanisms. Third, we hope to resolve through a beat frequency technique fine structure splitting in the 3 to 30 MHz region.

This program will require an IR detector with a rise time of 100 nsec permitting detection of difference frequencies as large as 30 MHz. The Teratron mode spacing is three times this frequency. But because of mode pulling effects of cavity resonance the frequency of oscillation in a laser is given by

$$f_0 = f_c + (f_m - f_c)\frac{\Delta f_c}{\Delta f_m} \quad ,$$

where f_c and Δf_c are the cavity frequency and linewidth, and f_m and Δf_m are the molecular transition frequency and linewidth. For example, two closely-spaced molecular transitions will yield laser oscillations at slightly different frequencies for the same laser mode

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number. The difference frequency between these oscillations then yields
information about the cavity mode and the two molecular transitions.
Thus it might be possible to study in laser emissions small field Zeeman
splittings; the hyperfine structure arising from magnetic interactions
between the electronic angular momenta and the magnetic moment of the
N nucleus; or spin doubling where the doubling is very small or is
narrowed during spin uncoupling by the magnetic field.

Another kind of question is apparent. Studies of He-Ne and Ar
lasers show a quenching of laser action at kilogauss fields and/or
high pressures; this quenching is associated with appearance of a ring-
shaped cross-sectional gain profile and radiation-trapping effects.[11]
The nature of this effect in the CN system needs to be studied.

The experimental effort stands as follows. The Teratron cyanide
laser arrived in October and is being checked out. Design work is
underway on a pulsed magnetic field of intermediate kilogauss strength;
as line shifting, quenching, and field homogeneity effects are estab-
lished the advisability of a steady state 10 kG or greater field will
become clear. The 100-nsec-response IR detector is being acquired.
Auxiliary equipment needed to resolve temporally the IR inversion kinetics,
to resolve the laser emission frequencies, and to carry out beat fre-
quency experiments is being studied.

REFERENCES

1. G. W. Chantry, H. A. Gebbie, and J. E. Chamberlain, Nature **205**, 377 (1965).

2. H. P. Broida, K. M. Evenson, and T. T. Kikuchi, J. Appl. Phys. **36**, 3355 (1965).

3. L. E. S. Mathias, A. Crocker, and M. S. Wills, Electronic Letters **1**, 45 (1965).

4. H. Steffan, J. Steffan, J. F. Moser, and F. K. Kneubühl, Phys. Letters **20**, 20 (1966).

5. H. Steffan, J. Steffan, J. F. Moser, and F. K. Kneubühl, Phys. Letters **21**, 425 (1966).

6. F. A. Jenkins, J. K. Root, and R. S. Mulliken, Phys. Rev. **39**, 16 (1932).

7. H. Steffan, P. Schwaller, J. F. Moser, and F. K. Kneubühl, Phys. Letters **23**, 313 (1966).

8. R. L. Brown and H. P. Broida, J. Chem. Phys. **41**, 2053 (1964).

9. H. E. Radford and H. P. Broida, Phys. Rev. **128**, 231 (1962).

10. C. H. Townes and A. L. Schawlow, Microwave Spectroscopy (McGraw-Hill, 1955), p. 286.

11. I. Gorog and F. W. Spong, Appl. Phys. Letters **9**, 61 (1966).